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Dr. Jeremy T. Busby Fuel Cycle and Isotopes Division P.O. Box 2008 – MS-6138 Oak Ridge, TN 37831-6138 Telephone: (865) 241-4622 Fax: (865) 241-3650 E-mail: busbyjt@ornl.gov

May 24, 2013

Dr. C. Smith RISMC Pathway Lead Idaho National Laboratory 2525 Fremont Avenue Idaho Falls, ID 83415

Subject: Completion of LWRS Level 3 Milestone (M3LW-13OR0704032) "Deliver status report describing implementation of changes to the EONY model to account for extended service conditions and late blooming phases for reactor pressure vessel steels", due 5/24/2013.

Dear Sir;

This letter is to inform you of completion of Milestone M3LW-13OR0704032— "Deliver status report describing implementation of changes to the EONY model to account for extended service conditions and late blooming phases for reactor pressure vessel steels" for the Enabling Methods and tools workpackage within the LWRS Risk-Informed Safety Margin Characterization Pathway. This work is in support of the larger effort to develop the GRIZZLY tool for simulation of materials aging in key reactor structures.

As noted in previous reports, there is a great deal of uncertainty in the amount of high dose, high fluence embrittlement that is may occur under irradiation in extended service scenarios. If new mechanisms of embrittlement are confirmed for extended service conditions, modeling and simulation tools will be required to evaluate operating limits and accident scenarios. The primary objective of GRIZZLY effort within the Light Water Reactor Sustainability Program is to develop robust models that can estimate RPV embrittlement and vessel integrity during PTS accidents at high fluence (φt) to at least 10²⁰ n/cm² (>1 MeV) pertinent to plant operation of some pressurized water reactors (PWR) for 80 full power years. This report updates progress on extensions that are required to the Eason, Odette, Nanstad and Yamamoto (EONY) model of RPV embrittlement for high fluence conditions and the implementation of these models into GRIZZLY.

The attached pages first review the suggested upgrades to the EONY model as described in an earlier report (M3LW-13OR0401052, Deliver assessment of needs to extend EONY (Eason, Odette, Nanstad, and Yamamoto) model and needs for implementation into Grizzly). Additional analysis of those needs is provided along with key steps, recent

progress and experimental status. Finally, supporting efforts in other programs and integration steps within GRIZZLY are discussed.

If you have any comments or questions, please contact me. Thank you.

Sincerely,

Dr. Jeremy T. Busby Senior Research Scientist Oak Ridge National Laboratory

CC: D. Williams, ORNL

T. Rosseel, ORNL

B. Wirth, Univ. of Tennessee and ORNL

R. Martineau, INL

Status report describing implementation of changes to the EONY model to account for extended service conditions and late blooming phases for reactor pressure vessel steels

J.T. Busby and B.D. Wirth, Oak Ridge National Laboratory University of Tennessee, Knoxville

1. BACKGROUND

The reactor pressure vessel (RPV) represents the primary boundary for release of coolant and radioactivity in an accident scenario. The regulations that govern the operation of commercial nuclear power plants require conservative margins of fracture toughness, both during normal operation and under accident scenarios. During reactor operation, leakage of neutrons from the reactor core pass through the RPV resulting in irradiation embrittlement that can severely degrade the RPV fracture toughness, with the degree of toughness loss dependent on the radiation sensitivity of the materials. The available embrittlement predictive models, e.g. [1,2], and our present understanding of radiation damage are not fully quantitative, and do not treat all potentially significant variables and issues, particularly considering extension of operation to 80 years.

For example, there are sparse or nonexistent data at high fluences, for long times, and the question of embrittlement at higher fluence over longer times create large uncertainties for embrittlement predictions [3,4]. This issue is directly related to life extension with the number of plants requesting license extension to 60y and those expected to request 80y. Moreover, there is now experimental evidence that so-called "late blooming phases"(rich in nickel and manganese) could produce an effect that could have serious implications to RPV life extension [5]. An example of late blooming phases from an operating power reactor is shown in Figure 1 below. The Ni-Mn-Si particles observed in a low-Cu weldment correlate with the very high measured temperature transition shifts. Additional research is required to understand the compositional, flux, fluence, and temperature dependencies. Some of these issues are being addressed under the experimental elements of the LWRS Materials Aging and Degradation Pathway. As well, the model requires improvements that could provide a rationalization for the possibility of lateblooming phases, which have been long postulated and are now being observed in RPV steels [6].

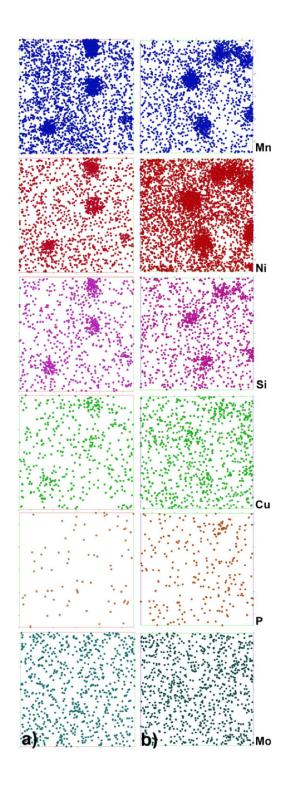


Figure 1: Atom maps of the low copper weld metal from the Ringhals R4 Unit neutron irradiated to fluences of (a) 3.30×10^{23} and (b) 6.03×10^{23} n/m² (E > 1 MeV) showing the solute distribution in the precipitates [5].

As noted in previous reports [7], there is a great deal of uncertainty in the amount of high dose, high fluence embrittlement that may occur under irradiation in extended service scenarios. If new mechanisms of embrittlement are confirmed for extended service conditions, modeling and simulation tools will be required to evaluate operating limits and accident scenarios. The primary objective of the GRIZZLY effort within the Light Water Reactor Sustainability Program is to develop robust models that can estimate RPV embrittlement and vessel integrity during PTS accidents at high fluence (φt) to at least 10²⁰ n/cm² (>1 MeV) pertinent to plant operation of some pressurized water reactors (PWR) for 80 full power years. This report updates progress on extensions that are required to the Eason, Odette, Nanstad and Yamamoto (EONY) model of RPV embrittlement for high fluence conditions and the implementation of these models into GRIZZLY.

2. RECOMMENDED DEVELOPMENT EXTENSIONS OF EONY MODEL

As discussed in [7] the EONY model can be improved in several ways that would maintain good agreement with surveillance data and improve the agreement with IVAR data, other test reactor data, and theoretical results. Preliminary improvements could be made using the 2004 surveillance database and the IVAR data, although newer surveillance data may also be available. The model extensions needs, current status, and future needs are listed below for each of the key areas.

2.1 Explicit Mn Effect

In the EONY model [1,2], the effect of Mn is mainly an implicit effect in the product form coefficients. Previously, an attempt during the modeling effort to include an explicit Mn effect was not successful, although understanding and incorporating the effects of Mn will be important for estimating the effects of late-blooming phases. The fitting form for the Mn effect and its interaction with Ni will need to be guided by theory and test reactor data, since a clear trend did not emerge from the large amount of scatter in the surveillance database.

In order to address this need, additional experimental data will be required including both commercial steels and single-variable model alloys with a range of Mn compositions under different irradiation conditions. This could be a significant experimental program by itself. Fortunately, two ATR-NSUF grants and two university programs are already providing these. The ATR-2 experiment includes 180 different alloys in the form of 1400 specimens. These include multi-purpose coupons, compact tension specimens, diffusion multiples, and sub-sized tensile specimens. This experiment will be carried to very high fluences and support this particular need for GRIZZLY development.

However, a number of delays in operation of the ATR have pushed the completion of the ATR-2 irradiation campaign deep into 2013. Chief among these is the Powered Axial Locator Mechanism (PALM) cycle, which is occasionally performed by the ATR for complex transient testing and which can simulate multiple start-up and shutdown cycles of tests for fuels and materials. The PALM tests normally last from a few hours to a couple of days, but some

experiments must be removed from the reactor when a PALM test is performed. This is the case for the ATR-2 capsule, which was removed in anticipation of the PALM cycle experiment. Thus the capsule irradiation capsule will not be complete, and the capsule removed from the reactor until September 2013. As data is analyzed it will be considered for the update of EONY.

High fluence irradiations of commercial steels will also be required to validate individual elements of embrittlement correlations. These are being collected as part of the LWRS Materials Aging and Degradation Pathway tasks. Materials from Ringhals 3 and 4, Ginna, Nine Mile Point, and Palisades are actively being harvested and analyzed. A list of other high value capsules has also been assembled for future harvesting including Kewaunee and Palisades. Early data from Ringhals (show above) highlights the importance of this EONY model update.

This effect and EONY update will be explored in more detail in the coming years as additional single-variable data becomes available.

2.2 Revised effective fluence form

The most obvious departure of the EONY model [1,2] when comparing with the IVAR database and theory is in the effect of neutron flux, specifically the breakpoint above which the effect of flux is not statistically significant in the surveillance data. This inconsistency is an artifact of the fitting constraints utilized in fitting the initial EONY model, and can be fixed by simply considering the clear trend from non-surveillance data when calibrating the effective fluence function. The effective fluence could be fit to the combined surveillance and IVAR data or to just the surveillance data, using a constraint on slope at higher flux based on the IVAR data.

As above in section 2.1, additional experimental data will be required including both commercial steels and single-variable model alloys with different flux and fluence conditions. The ATR-NSUF programs mentioned above will also be a key resource in this step. The high fluence capsules are of particular value although will not be available until the end of this calendar year. As data is analyzed it will be considered for the update of EONY.

2.3 Temperature effect in Matrix Feature (MF) and Copper Rich Precipitate (CRP) terms

During EONY calibration, an attempt to include a temperature term in the CRP part of the model was not successful, possibly because it was added after a temperature effect had been calibrated in the MF term. Analogous to the Mn effect, it appears that the residual effect of temperature in higher Cu surveillance data is small when the MF term temperature effect has already been calibrated. Three different strategies to explore this feature were identified including estimate the temperature effects for MF and CRP features from non-surveillance data, apply the same temperature effect to both terms in the model, since the MF temperature effect already is applied to all materials or to calibrate the temperature effect in the MF term to just the low Cu materials, even though the MF term applies to all Cu levels, then hold that MF temperature effect fixed and calibrate a CRP temperature effect to just the higher Cu materials.

As in the two previous sections, the ATR-2 experiment with University of California Santa Barbara will be essential in refining these terms. As data is analyzed it will be considered for the update of EONY.

Access to a broader range of data from surveillance coupons and service conditions will serve on route to completing analysis of the temperature effect. Fortunately, the FAVOR project also includes a database incorporating all surveillance data from all US plants. Case studies can be made for the highest known fluences for given plant geometries and compared to known data sets.

Full access to these data sets is currently being negotiated. A contract with Dr. Eason to perform the analysis described above is also under development and is expected to be in place by the end of June 2013.

2.4 Add third embrittling feature to EONY model

As noted in the previous report, a third feature was considered. The JOM paper by Odette and Nanstad [4] describes preliminary work on modeling the formation of unstable matrix defects (UMD) that was done in conjunction with the EONY model, which are important aspects of the irradiated microstructure at very high neutron flux. These features, in addition to the extent in which these may contribute to or mediate the formation of CRP or late blooming phases need to be considered in the model to accurately predict long-term embrittlement behavior.

However, upon further analysis and consideration, it is possible that inclusion of such a UMD term may be premature. Subsequent analysis by Eason and Odette [8,9] suggests that the behavior of unstable matrix defects (UMDs) perhaps does not belong in the model as a third term, but rather should be considered as part of deciding "how to use but not abuse" the available higher flux data.

As noted by Eason [8], this can be interpreted as two key issues. First, the understanding of UMDs is still evolving, and it would be premature to build into the EONY model an incomplete understanding that may later be deemed incorrect. Also, the fact that UMDs form and change under the higher flux fields of test reactors (but not so much in power reactors) means that UMDs should be considered in choosing relevant higher flux data, but not necessarily model the details. The timing for deciding how or whether to include UMDs as a model feature would be better after the ATR-2 results are completed and analyzed.

Given the additional input and thought on the incorporation of this feature, no additional work on the UMD term is currently planned. However, this analysis, in combination with the experimental results presented in Figure 1 of this report, clearly identifies the addition of a term describing the embrittlement contributions of late-blooming phases as a priority addition to the model. Additional thought and consideration on how to add this term is needed and will be pursued in the coming months. Theoretical development and more high fluence data, special composition observations, etc are needed before EONY may properly add LBPs, and their potential importance to power reactors at long life is such that correct model implementation is

essential. A first step toward future LBP implementation is improving the Mn, flux, and temperature characterization in the EONY model, which can be done sooner, but it would be premature to promise adding the LBP feature at this time.

3. GRIZZLY Implementation and upgrades

The EONY model for RPV embrittlement has been implemented into GRIZZLY, as documented in the initial assessment report [10]. The most pressing next steps within the GRIZZLY implementation effort are to introduce multiple plate and weld materials throughout the reactor pressure vessel, and to evaluate the effect of material inhomogeneity on stress profiles that can impact structural integrity during PTS scenarios. Several steps must be taken in the near term to accomplish relevant geometries including incorporation of actual weldments and heat-affected zones, and most importantly, inclusion of the vessel liner. This work is ongoing between INL, ORNL and the University of Tennessee.

During the past six months, additional work has been performed to develop an interface between the GRIZZLY and FAVOR codes. By coupling these two modeling tools, additional validation is possible. More importantly, by utilizing tools such as fracture mechanics in FAVOR and the 3D capability of GRIZZLY. This interface has been completed and thermal / stress benchmarking between GRIZZLY and FAVOR thermal and stress analysis was successful for 1-D problems. Additional locations in the FAVLOAD interface where further modifications are required to read in GRIZZLY output have been identified and coding to modify FAVLOAD is complete.

Next steps in the development of GRIZZLY have also been discussed and identified. In the coming months, several key steps will be initiated, in addition to finalizing the incorporation of the liner into the model (being performed under a different work package). Evaluating the three-dimensional effects around nozzles with very low dose irradiation and residual stresses, could be important under extended service conditions as nozzles will begin to see appreciable does. This complex component and geometry will require 3-D simulations. In addition, this 3-D capability will also be used to explore the effects of 3-D crack/flaw orientation effects using the Belgian Doel-3 reactor as a prototypic geometry and motivation.

4. SUPPORTING MODELING ACTIVITIES

As part of the other elements of GRIZZLY, there is some effort to simulate nucleation of late blooming phases in RPV steels. In recent months, INL has run a validation case looking at the diffusion behavior of a single vacancy in body-centered cubic iron modeled with molecular dynamics simulations and compared the results to atomic kinetic monte carlo results. This code will be expanded to multi-alloy systems as in reactor pressure vessel steels in the future. Clearly, understanding nucleation would augment broader EONY and GRIZZLY needs. Additional coordination and discussion with this effort will be pursued in the coming months.

In addition to LWRS activities, there is ongoing research as part of a recent NEUP program that is relevant and supporting to the update of EONY and GRIZZLY. Recent work by the University of California Santa Barbara and University of Wisconsin has direct impact on this effort. In their ongoing effort, modeling work has been motivated by the detection of Fe in Ni-Mn-Si rich precipitates in RPVs (so-called Late Blooming Phases). This observation raises an important question as understanding the nature and composition of these particles, which may be stoichiometric G-phase Mn₆Si₇Ni₁₆, based on limited microscopy analysis. Given this apparent discrepancy, it is essential to derive terms for model estimation. Further, due to the small size of these precipitates, the detected Fe might be just the matrix rather than solutes in precipitates. To test whether Fe is soluble in these precipitates, the UCSB/UW team are calculating the heat of solution and solubility of Fe in these phases with ab initio methods (VASP) and have completed this study for G-phase. Results show essentially no Fe solubility (less than 0.2% when replacing Mn and 0.02% when replacing Ni) under typical late blooming phase and RPV steel conditions. Further work is ongoing to study thermodynamics and kinetics of G-phase like precipitates in these systems. Monthly coordination calls (and webinars) have been established between these efforts to provide additional support to the EONY and GRIZZLY updates.

5. SUMMARY

As noted above, several key changes to the EONY model were recommended, including

- Introduce an explicit dependence on the alloy Mn content in the transition temperature model:
- Revise the effective fluence dependence to better match test reactor databases;
- Revise the irradiation temperature dependence with a more physically realistic description; and
- Add a third embrittling feature.

Additional analysis and work on these has resulted in a prioritization and a more detailed examination of needs. The first three will all rely on additional experimental data that is in progress. Further work on the irradiation temperature dependence utilizes existing surveillance data and contracts are being negotiated.

Supporting work as part of a NEUP program and another work package are being coordinated to further bolster EONY and GRIZZLY updates.

6. REFERENCES

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